# NAVAL POSTGRADUATE SCHOOL

## MONTEREY, CALIFORNIA

# THESIS

## CHARACTERIZATION OF A CONTINUOUS WAVE LASER FOR RESONANCE IONIZATION MASS SPECTROSCOPY ANALYSIS IN NUCLEAR FORENSICS

by

Sunny G. Lau

June 2015

Thesis Advisor:                                         Craig Smith
Co-Advisor:                                             Dragoslav Grbovic

**Approved for public release; distribution is unlimited**

THIS PAGE INTENTIONALLY LEFT BLANK

| REPORT DOCUMENTATION PAGE | | | *Form Approved OMB No. 0704–0188* |
|---|---|---|---|

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instruction, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202–4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704–0188) Washington DC 20503.

| 1. AGENCY USE ONLY *(Leave blank)* | 2. REPORT DATE<br>June 2015 | 3. REPORT TYPE AND DATES COVERED<br>Master's Thesis | |
|---|---|---|---|
| **4. TITLE AND SUBTITLE**<br>CHARACTERIZATION OF A CONTINUOUS WAVE LASER FOR RESONANCE IONIZATION MASS SPECTROSCOPY ANALYSIS IN NUCLEAR FORENSICS | | **5. FUNDING NUMBERS** | |
| **6. AUTHOR(S)** Sunny G. Lau | | | |
| **7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)**<br>Naval Postgraduate School<br>Monterey, CA 93943–5000 | | **8. PERFORMING ORGANIZATION REPORT NUMBER** | |
| **9. SPONSORING /MONITORING AGENCY NAME(S) AND ADDRESS(ES)**<br>N/A | | **10. SPONSORING/MONITORING AGENCY REPORT NUMBER** | |

**11. SUPPLEMENTARY NOTES** The views expressed in this thesis are those of the author and do not reflect the official policy or position of the Department of Defense or the U.S. Government. IRB Protocol number ____N/A____.

| 12a. DISTRIBUTION / AVAILABILITY STATEMENT<br>Approved for public release; distribution is unlimited | 12b. DISTRIBUTION CODE |
|---|---|

**13. ABSTRACT (maximum 200 words)**

The application of resonance ionization mass spectroscopy (RIMS) to nuclear forensics involves the use of lasers to selectively ionize elements of concern. While current systems incorporate pulsed lasers for analysis of debris from nuclear detonation, the possibility exists to consider using continuous wave, or CW lasers

RIMS has the potential to provide rapid quantification of isotope ratios of important elements in debris from nuclear detonation. The current approach to ionize uranium and plutonium uses three Ti-Sapphire pulsed lasers capable of a fundamental wavelength range of 700–1000 nm to perform ionization in three steps. This thesis evaluates the use of COTS CW laser to replace one of the pulsed lasers exciting the second resonance step of plutonium near 847.282 nm.

The thesis research consists of three elements: (1) completion of an initial feasibility study to determine the viability of the COTS CW alternative, (2) identification and acquisition of a candidate COTS laser, and (3) testing to evaluate the critical laser parameters necessary to achieve high precision isotope ratio measurements including the stability over time of the mean wavelength, bandwidth and spectral mode purity.

This narrower bandwidth COTS CW laser may enable simpler operations, greater robustness and better utilization of the available laser irradiance.

| **14. SUBJECT TERMS**<br>RIMS, resonance ionization, mass spectroscopy, nuclear forensics, continuous wavelength laser, CW, plutonium, COTS, isotope, characterization | | | **15. NUMBER OF PAGES**<br>57 |
|---|---|---|---|
| | | | **16. PRICE CODE** |
| **17. SECURITY CLASSIFICATION OF REPORT**<br>Unclassified | **18. SECURITY CLASSIFICATION OF THIS PAGE**<br>Unclassified | **19. SECURITY CLASSIFICATION OF ABSTRACT**<br>Unclassified | **20. LIMITATION OF ABSTRACT**<br>UU |

THIS PAGE INTENTIONALLY LEFT BLANK

# CHARACTERIZATION OF A CONTINUOUS WAVE LASER FOR RESONANCE IONIZATION MASS SPECTROSCOPY ANALYSIS IN NUCLEAR FORENSICS

Sunny G. Lau
Lieutenant Commander, United States Navy
B.S., Oregon State University, 2003
M.S., Naval Postgraduate School, 2004

Submitted in partial fulfillment of the
requirements for the degree of

**MASTER OF SCIENCE IN APPLIED PHYSICS**

from the

**NAVAL POSTGRADUATE SCHOOL
June 2015**

Author:                  Sunny G. Lau

Approved by:       Craig Smith
                       Thesis Advisor

                       Dragoslave Grbovic
                       Co-Advisor

                       Andres Larraza
                       Chair, Department of Physics

THIS PAGE INTENTIONALLY LEFT BLANK

# ABSTRACT

The application of resonance ionization mass spectroscopy (RIMS) to nuclear forensics involves the use of lasers to selectively ionize elements of concern. While current systems incorporate pulsed lasers for analysis of debris from nuclear detonation, the possibility exists to consider using continuous wave, or CW lasers

RIMS has the potential to provide rapid quantification of isotope ratios of important elements in debris from nuclear detonation. The current approach to ionize uranium and plutonium uses three Ti-Sapphire pulsed lasers capable of a fundamental wavelength range of 700–1000 nm to perform ionization in three steps. This thesis evaluates the use of COTS CW laser to replace one of the pulsed lasers exciting the second resonance step of plutonium near 847.282 nm.

The thesis research consists of three elements: (1) completion of an initial feasibility study to determine the viability of the COTS CW alternative, (2) identification and acquisition of a candidate COTS laser, and (3) testing to evaluate the critical laser parameters necessary to achieve high precision isotope ratio measurements including the stability over time of the mean wavelength, bandwidth and spectral mode purity.

This narrower bandwidth COTS CW laser may enable simpler operations, greater robustness and better utilization of the available laser irradiance.

THIS PAGE INTENTIONALLY LEFT BLANK

# TABLE OF CONTENTS

THIS PAGE INTENTIONALLY LEFT BLANK

# LIST OF FIGURES

THIS PAGE INTENTIONALLY LEFT BLANK

# LIST OF TABLES

THIS PAGE INTENTIONALLY LEFT BLANK

# LIST OF ACRONYMS AND ABBREVIATIONS

| | |
|---|---|
| CB | Conduction band |
| COTS | Commercial-off-the-shelf |
| CW | Continuous wave |
| DTRA | Defense Threat Reduction Agency |
| IR | Infrared |
| LED | Light Emitting Diode |
| MOPA | Master Oscillator Power Amplifier |
| PRF | Pulse repetition frequency |
| RIMS | Resonance Ionization Mass Spectrometry |
| TEM | Transverse Electric Magnetic |
| UV | Ultraviolet |
| VB | Valence band |

THIS PAGE INTENTIONALLY LEFT BLANK

# ACKNOWLEDGMENTS

THIS PAGE INTENTIONALLY LEFT BLANK

# I.    INTRODUCTION

## A.    BACKGROUND AND MOTIVATION

In the current world with the increasing availability of nuclear materials such as uranium and plutonium, it is becoming easier for our adversaries to do harm through dirty bombs or improvised nuclear devices. Using the methods of nuclear forensics, the origins of such nuclear materials can potentially be traced back to a country or particular source [1]. A nuclear forensics technique that can distinguish and quantify the isotopes of interest in a debris sample without the use of complex and time-consuming chemistry techniques is called Resonance Ionization Mass Spectrometry (RIMS) [2]. The RIMS approach uses lasers operating at a specific ionization wavelength to selectively ionize the atoms of uranium or plutonium through particular electron transitions; the ions are then subjected to a mass spectrometry process to provide a quantitative analysis of the isotopic composition of the debris sample in question (see Figure 1) [3], [4].
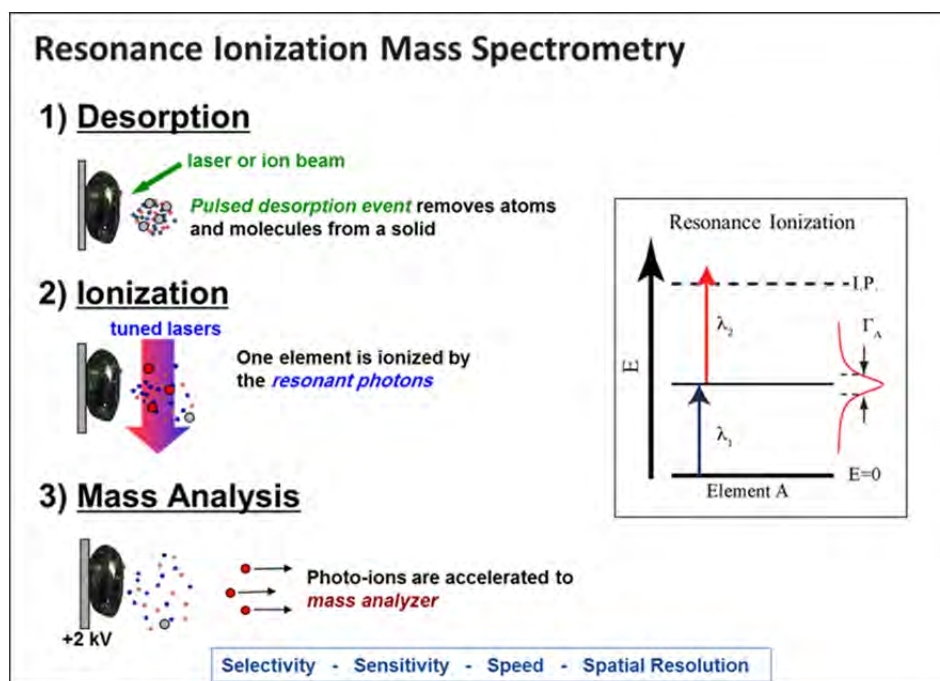


Figure 1.    RIMS selective ionization and mass analysis process, from [2]

This thesis describes the research conducted to study the operational parameters and performance characteristics required of the laser(s) used in the RIMS analysis of plutonium to consider the possibility that a commercial-off-the-shelf (COTS) alternative laser may offer a viable cost effective and portable solution compared to the existing laboratory laser systems in place today. The laboratory RIMS system is capable of both uranium and plutonium isotope discrimination. Plutonium isotopes were selected for this research and will be referred to in subsequent discussions.

### 1.    Current RIMS Laser Setup

During the final ionization stage of RIMS, "excited atoms are photo-ionized from the intermediate electronic state by photons from an additional laser tuned to provide enough energy to place the excited atoms at or over the ionization threshold for that element" [5]. The laser system that produces the photons for ionization is comprised of three tunable lasers (Ti:Sapph, $Al_2O_3$ crystal doped with Ti) pumped by a commercial lasers (Nd:YLF, $LiYF_4$ crystal doped with Nd) see Figure 2 [2]. Three Ti-Sapphire lasers are used to ionize three electronic levels in both $^{235}U$, $^{238}U$, and $^{239}Pu$, $^{240}Pu$. The wavelength for the different levels ranges from: $\lambda_1 = $ ~415–420 nm, $\lambda_2 = $ ~829–847 nm, $\lambda_3 = $ ~722-767 Nm [2]. Notice the wavelength spreads in $\lambda_1$ and $\lambda_2$ of uranium and plutonium are less than 20nm, suggesting a tunable wavelength laser may be suitable for excitation for the first two steps for all four isotopes of primary interest.
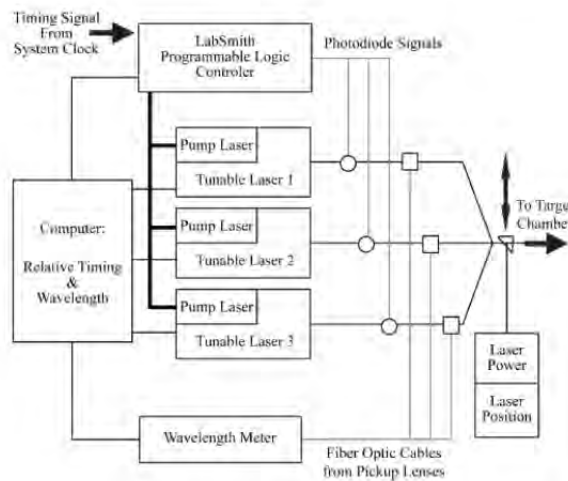


Figure 2.    Current RIMS pump and ionization laser setup, from [2]

This research is focused on considering possible replacement solutions for at least one of the pulsed lasers; however, a solution for one laser may well prove to be applicable to two or perhaps three of the excitation/ionization lasers.

## B.     LASERS AS AN ENERGY SOURCE

Lasers are ideal for RIMS applications because they are coherent light sources. (The term "light" in this thesis will be defined as any electromagnetic energy ranging from UV to IR). Natural light sources have linewidths ($\Delta\lambda$) ranging from 250–2500 nm (or $\Delta\lambda \approx 2250$ nm), LED light sources [6] are much narrower ranging from $\Delta\lambda \approx 30 - 50$ nm. A monochromatic laser light source, which emits light that is nearly pure in color [7] can exhibit linewidths of $\Delta\lambda \approx 10^{-8}$ nm which are substantially narrower than a standard laser's linewidth (see Figure 3 for a comparison of a LED to a laser linewidth). Such unique characteristics of lasers are based on their coherence.
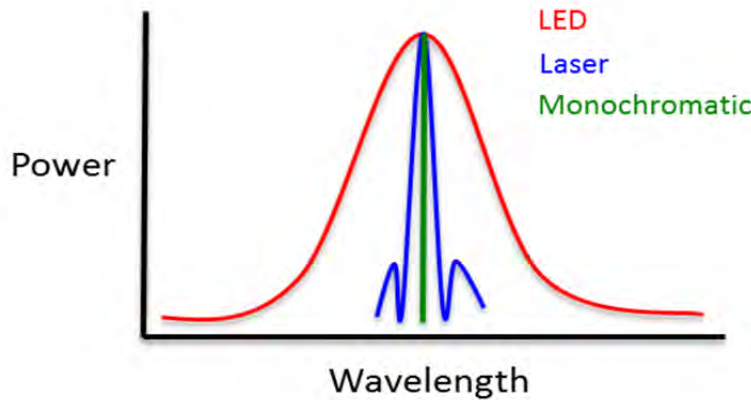


Figure 3.     Linewidth ($\Delta\lambda$) comparison of LED, laser and monochromatic light

### 1.     Temporal and Spatial Coherence

There are no *perfectly* coherent light sources, but lasers are as close as can be achieved. Coherence can be understood as "a measure of phase correlation in a light source at different locations and times" [8]. Incoherence is when the phase correlation is random. There are two types of coherence, *temporal* and *spatial*, each of which affects the quality and usability of a beam.

"*Temporal* coherence considers the correlation in phase between *temporally* distinct points of a radiation field along its line of propagation" [8]. Temporal coherence relates to the monochromaticity (i.e., pureness of "color") and the corresponding linewidth ($\Delta\lambda$) of a laser source. From representation of the harmonic wave train with center frequency ($\omega_o$) and lifetime of $\tau_o$ the relation is given:

$$\Delta\omega = \frac{2\pi}{\tau_o}$$

meaning that as the lifetime approaches infinity the frequency length ($\Delta\omega$) approaches zero or approaches a single frequency $\omega_o$. Based on a single sinusoidal wave train with lifetime ($\tau_o$) the following relationships can also be found [8], [9].

$$\Delta\lambda \cong \frac{\lambda^2}{l_t} \ \ where \ l_t = c\tau_o$$

indicating that a longer coherence length ($l_t$) commonly associated with a continuous laser results in a narrower natural linewidth ($\Delta\lambda$), and a long coherence time ($\tau_o$) is associated with a wave that takes a longer in time to drift in phase.

"*Spatial* coherence considers the correlation in phase of *spatially* distinct points of a radiation field" [8]. The propagation of two points at one wave front can be correlated to a different wave front if it is spreading or diverging (Figure 5) [8].
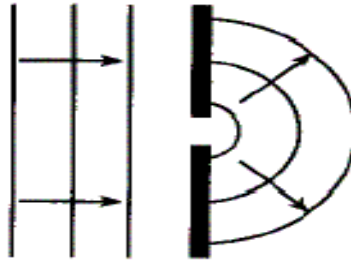


Figure 4.    *Left* - Parallel propagation of two points
*Right* - Diverging propagation of same two points, from [8]

The overall spreading (θ) of a beam can be calculated by: $\theta \sim \dfrac{\lambda}{D_c}$ where $\lambda$ is wavelength and $D_c$ is aperture diameter. When points along a wave front do not exactly correlate in a lasing cavity the off-axis transverse mode can self-replicate (see Figure 5) [9], resulting in multiple concentrations of intensities.

Spatial coherence measures the degree of correlation among the transverse modes within a lasing cavity. The notation to define transverse modes is called Transverse Electric and Magnetic (TEM) where $TEM_{00}$ is the fundamental mode [8]. Higher modes will result in shapes other than a single spot, examples of higher modes are shown in Figure 5.
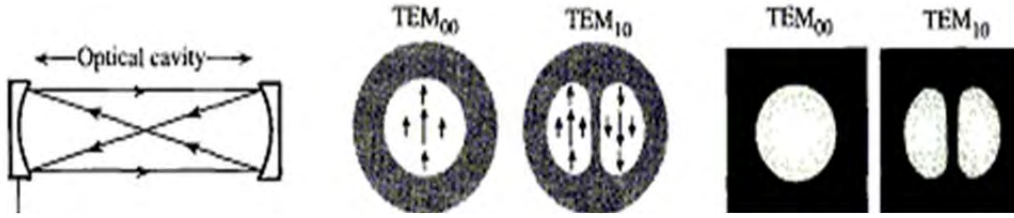


Figure 5.    Transverse Electric Magnetic (TEM) mode examples, from [9]

Where temporal coherence relates to the monochromaticity and linewidth ($\Delta\lambda$) of a beam, spatial coherence relates to the modes (shape) of a beam. It can be said that a laser with good coherence will inevitably have narrow linewidth, near single lasing frequency, and sharp interference fringe(s) [8].

Characterization and selection of a laser can be confusing. Terms such as Free Electron, Ti-Sapphire, Continuous Wave are all used to describe types of lasers. To minimize confusion lasers can be broken down to three characterization topics based on; (1) the material (gain medium) used to achieve lasing; (2) the laser output power mode; and (3) the laser wave properties.

## C.    SUMMARY AND OVERVIEW OF THE THESIS

In summary, the RIMS system relies on lasers for the ionization of atoms. Such lasers are capable of ionization if they are tuned to a particular wavelength, have a

narrow linewidth, and operate in a single mode. In Chapter II, laser definitions and characterizations is broken down into lasing material, laser power type, and laser wave properties. Chapter III discusses the initial feasibility study in COTS alternatives, then the identification, description, and breakdown of the candidate laser. Chapter IV discusses the initial testing of the critical parameters necessary for successful ionization. Chapter V reviews the findings with the candidate laser and provides recommendations for future work.

## II.     LASER CHARACTERIZATION FROM THEORY

### A.     THE LASER GAIN MEDIUM

There are various gain media available to achieve a desired wavelength laser. Some includes the use of chemicals, and dyes which are difficult to store, and transport and are susceptible to molecular inconsistencies. A second option is that of the semiconductor laser, a technology now widely available and used in everyday electronics such as DVD/Blue-Ray players, and fiber optic communications. Meeting the goal of the RIMS laser alternatives project, to have a cost effective, portable and stable laser a semiconductor laser was selected.

### 1.     Light Emission through a Semiconductor

A semiconductor laser is based on the *Pauli exclusion principle*, which states that restricts electron transitions from one energy state to another and defines the permissible process of the emission or absorption of photons. The band gap energy ($E_g$) necessary to transition from the valence and conduction band (VB) to the conduction band (CB) is represented by $E_g \approx h\nu$. This is also the energy that is released when an electron transitions from the conduction band back to the valence band. The semiconductor physical principles behind a LED and a laser diode are similar, and the process of creation of a photon ($h\nu$) is depicted in Figure 6 [7].
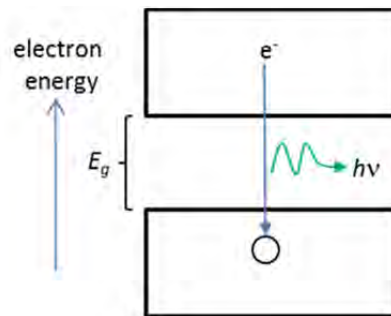


Figure 6.     Photon emission from electron-hole pair

## 2. Light Emitting Diodes (LED) and Diode Lasers

In addition to achieving a band gap energy, a diode's ability to continue recreating electron-hole pairs and emitting photons is achieved by a *forward biased* p-n junction. A p-n junction is created by doping (addition of impurities) to create semiconductor materials with higher concentration of positive (p) charge carriers or higher concentration of negative (n) carriers. When the p-n materials are brought together and voltage is applied to the p-side (*forward biasing*) the energy gap between the p and n side decreases (see Figure 7).
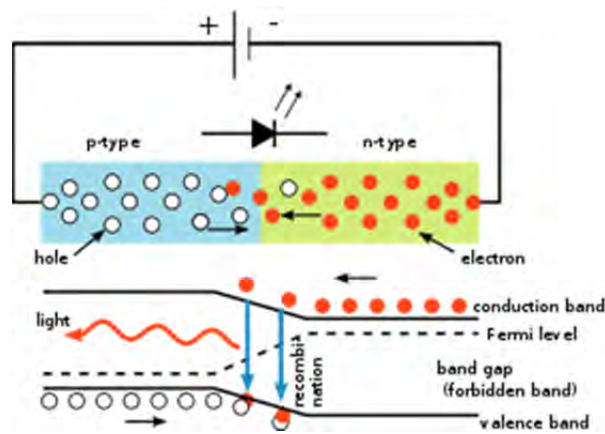


Figure 7.    Forward Biasing, from [6]

This smaller gap allows the diffusion of electrons from the n region to the p region, creating a current and leading to the emission of light (photons) when electrons (negative charge carriers) recombine with holes (positive charge carriers). This is how light emission begins in an LED or a laser diode. In order to progress from light emission to laser emission, the light needs to be amplified. The amplification can be accomplished by carefully positioned mirrors to pass the light back through the semiconductor material to create stimulated emission (resulting in similar wavelength, phase and polarization) [9], [10].

A semiconductor laser gain medium may be composed of gallium arsenide (GaAs) which is highly effective for efficient light emission because it uses a direct energy bandgap process rather than an indirect bandgap where a portion of the energy is consumed in *phonon* action. Bandgap energy can be changed as a function of a ternary

8

alloy (e.g., $Al_xGa_{1-x}As$) which allows for different wavelength lasers. The bandgap is related by: $E_g=1.424 + 1.427x+0.041x^2$eV [9]. With little or no moving parts other than electron-hole pair diffusion, the semiconductor laser can be made very small and quite durable. This gain medium is relatively stable in comparison to others using exotic materials, gases and chemicals, thus making it a good option for use in a portable RIMS system.

## B.    THE LASER POWER AND MODES

The two common operating modes of a laser are the pulsed and continuous wave modes. The difference between these two modes of operation is the nature of their power output which is either pulsed or continuous. It is possible to use a continuous wave (CW) laser and turn it on and off quickly to convert it to a pulsed laser and in some applications a CW laser is even used to seed a pulsed laser. The defining property of a CW laser is that the output power is steady (when averaged) in comparison to the cavity lifetime or period energy which is stored in the gain medium [11]. For the RIMS application, the amount of power illuminating a debris sample needs to be well understood and controlled.

### 1.    Pulsed Power Lasers

A pulsed laser, which for the current nuclear forensics RIMS system is a Ti-Sapphire gain element laser, is used primarily due to its large operating bandwidth. It provides bandwidths that range from 700–1000 nm. It has good efficiency with outputs that are ~20–30% of pump input power [9]. This is ideal for RIMS systems that have the desire to test numerous elements with large ranges of excitation wavelengths, but costly and complex for if only a few selected elements such as uranium or plutonium are desired.

A very important and limiting characteristic of a pulsed laser is its ability to produce high peak power by storing and releasing the laser energy at timed intervals (Q-switching techniques) [9]. A pulsed laser operating in such a wide bandwidth results in short pulses, ($\Delta t_p$) of 0.5–500 ns are common [12].

Mode locking, a technique to lock multiple longitudinal modes in phase is used to obtain even narrower pulses [9]. Pulse duration is inversely proportional to the material bandwidth $\left( \Delta t_p \cong \dfrac{1}{\Delta \upsilon} \right)$ as described in the literature [9]. Relating frequency bandwidth ($\Delta v$) at a particular wavelength ($\lambda$) is through the equation: $\Delta \upsilon = \dfrac{c}{\lambda^2} \Delta \lambda$ . In order to compare pulsed and CW lasers several more parameters are introduced. The pulse repetition frequency (PRF or $v$) is:

$$PRF = \frac{1}{T} ; T \simeq N \Delta t_p$$

PRF relates the pulse duration to the number of passes within the laser cavity. The PRF of currently-employed RIMS Ti-Sapphire laser is ~1000 Hz [2].

A pulsed laser has a relatively high peak power ($P_{pk}$) related by $P_{pk} = \dfrac{E}{\Delta t}$ , for the RIMS laser with an energy (E) of ~1 mJ and $\Delta t$ of 20 ns the $P_{pk}$ is ~50 kW [13]. The calculated average power ($P_{avg}$) is ~1 Watt from the relation $P_{avg} = E \times PRF$ ; this is a much lower power than at its peak pulse and can be used to related to the average continuous wave laser power.

## 2.    Continuous Wave (CW) Power Lasers

Recent studies have shown that for RIMS applications, a CW laser can be tuned to frequencies applicable to multiple isotopes of a given element, limited primarily by the time dependence of ionization, and the laser parameters of power and pointing stability [13]. A continuous wave laser provides continuous uninterrupted output power at a constant frequency and wavelength ($\lambda$) related by: $f = \dfrac{c}{\lambda}$ .

This type of laser is mature and commonly used in fiber optic communications to transmit information through amplitude changes in the wave. A CW laser has also a characteristically narrower linewidth ($\Delta\lambda$) given by: $\Delta \lambda \approx \dfrac{\lambda^2 \Delta \upsilon}{c}$ and inversely a higher

10

frequency change *(Δν)* [8]. CW lasers, with a narrower linewidth than pulsed lasers can potentially provide additional insight into the hyperfine structures of isotopes. With the proper vibrational and temperature controls in place a CW laser should be able to achieve continuous wavelength, power and pointing stability with this narrower linewidth. Traditionally CW diode lasers have been known to be power output limited but modern amplification techniques have been able to preserve linewidth and wavelength while providing higher output powers up to several Watts.

## C.    LASER MODE STRUCTURE

Whether lasers are pulsed or CW they are associated with the laser wavelength (center), its linewidth (bandwidth), and the modes. A laser wavelength *(λ)* is commonly the center of a laser's linewidth *(Δλ)* denoted in nanometers (nm). The modes of a laser are the allowable wavelengths which, based on a cavity length can produce a standing wave. The number of modes (m) available in a cavity is defined by $m = \dfrac{2nL\Delta f}{c}$, where n is the refractive index in the cavity, L is the cavity length , $\Delta f$ is the bandwidth in frequency and only exist as a multiple (not a distribution) of the gain medium's emission wavelength. It is possible for a laser to contain multiple modes and subsequently lase at different wavelengths [6].
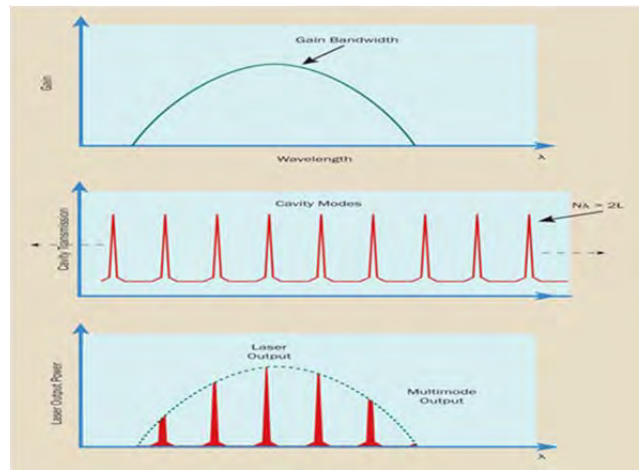


Figure 8.    Bandwidth, wavelength and modes (not equally spaced), from [12]

11

Figure 8, *top* shows the gain bandwidth of a laser or the bandwidth based on material that the laser can operate within. The *middle graphic* shows the various modes the laser can operate in based on the cavity length and properties. The *bottom graphic* shows the possible modes (not to scale) that can exist within a certain type of laser.

Single mode lasing (often used in high-precision scientific research) allows one mode of the light [14]. In the case of RIMS, a single mode laser (recall $TEM_{00}$) is preferred; multiple mode shapes are shown for comparison in Figure 9. From resonance ionization theory, ionization of a selected isotope is better predicted through a single known frequency within a single mode.
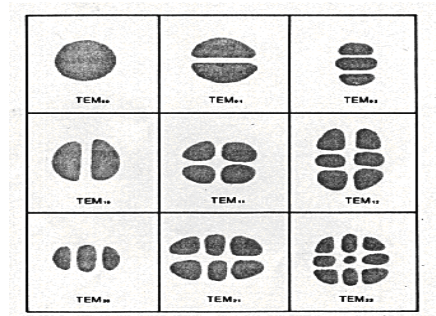


Figure 9.    Various TEM mode types, from [15]

Single mode can be characterized by a general Gaussian shape (see Figure 10) with a bell-shape curve and greatest intensity at the center, equally distributed circularly outwards.
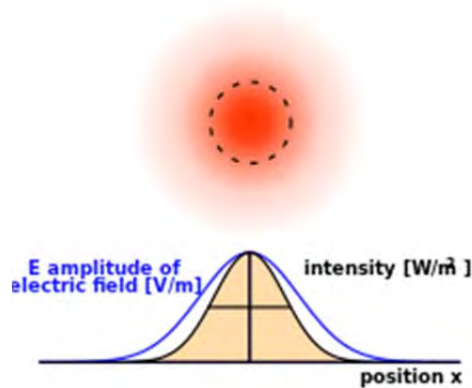


Figure 10.    General Gaussian distributed beam profile, from [16]

12

Although single mode lasing is preferred, multiple modes is a natural occurrence of lasers and should be understood where it exists and if it is to be mitigated. With this understanding a beam profile that deviates in shape from Gaussian may suggest multimode lasing.

## D.    THE LASER WAVE PROPERTIES

Useful characteristics of lasers are stability and relatively narrow linewidth *(Δλ)* at a desired wavelength; accomplished by mode, frequency stability and laser tuning [6]. The methods to control these parameters are wide and varying but control of the parameters is what is necessary for predictable RIMS techniques.

### 1.    Single Mode Lasing

Lasers will inherently have multiple modes as a result of constructive transverse interference. Single mode lasing assumes a narrow linewidth beam that only encompasses a single mode. It is a joint interaction of temporal and spatial coherence that results in single mode lasing. The interaction can be controlled in numerous ways and the most common method is through the control of the cavity length to select a single color from a polychromatic beam using gratings and angle adjustments. Methods employed to implement simultaneous temporal and spatial controls result in what is known as a *wavelength tunable laser* [17].

### 2.    Mode Frequency Stability

Control of the wavelength to single mode limits it to one wavelength; however, it remains possible to have multiple modes with a gain medium (see Figure 8). The mode frequency of a laser can change in time due to temperature changes (heating up) affecting the index of refraction and the speed of light within the cavity. Active methods to limit frequency fluctuations include use of a piezoelectric transducer controlled through a feedback loop from the desired frequency [6]. Mode frequency stability is important to understand in determining wavelength and shape.

THIS PAGE INTENTIONALLY LEFT BLANK

# III. FEASIBILITY, SELECTION AND ACQUISITIONS

## A. INTRODUCTION

The primary efforts in this thesis research include feasibility evaluation, selection/acquisition and testing of a candidate COTS laser for RIMS application. This Chapter describes the first two of these areas (feasibility evaluation, including definition of the characteristics that set the requirements for a RIMS CW laser, and selection/acquisition), while the following chapter addresses the testing completed in this research. In Section B, the known requirements on performance, availability and cost are considered. In Section C, the selected COTS laser is introduced and the physics broken down to demonstrate that it is capable of meeting the desired capabilities.

## B. COMMERCIAL-OFF-THE-SHELF (COTS) LASERS

The conditions considered in evaluating the feasibility of using COTS CW lasers for the RIMS application, in addition to required performance characteristics, were cost, availability, and ease of implementation. The characteristics of such an alternative laser include the need to meet the minimum wavelength properties to excite Pu at either $\lambda_1 = 420.760$ nm or $\lambda_2 = 847.282$ nm. The reason for requiring a CW laser with a power of 2 Watts was to ensure above 90% target element ionization (*left* Figure 11, red-dashed line). At 1 Watt, the irradiance of a pulsed wave laser was measured to produce $6.96 \times 1015$ W/m$^3$ irradiance. However, as low as 40% of that power was sufficient for >90% ionization (or $2.97 \times 1015$ W/m$^3$ irradiance) Since 2 W power on the CW laser produced sufficient irradiance, that level was chosen for our experiment (see Figure 11).

Plutonium Saturation as a Function of Laser Power

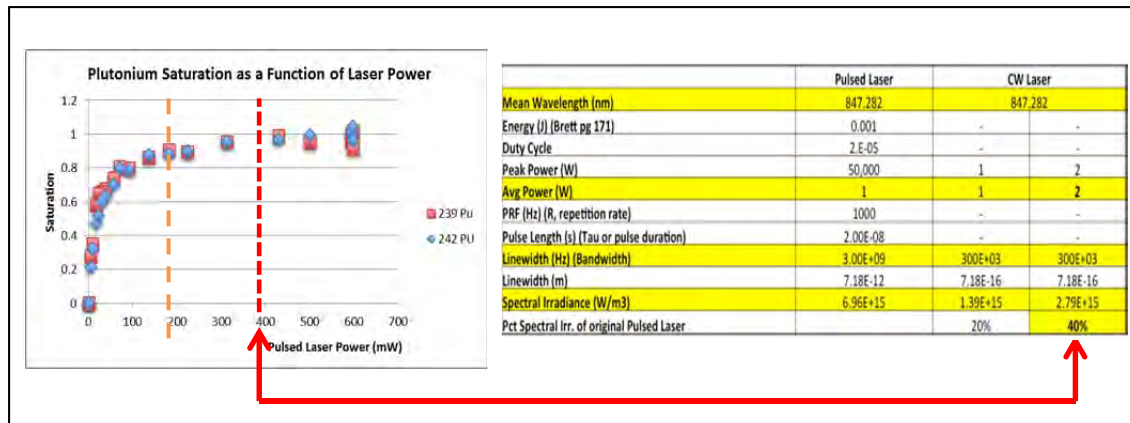| | Pulsed Laser | CW Laser | |
|---|---|---|---|
| Mean Wavelength (nm) | 847.282 | 847.282 | |
| Energy (J) (Brett pg 171) | 0.001 | - | - |
| Duty Cycle | 2.E-05 | - | - |
| Peak Power (W) | 50,000 | 1 | 2 |
| Avg Power (W) | 1 | 1 | 2 |
| PRF (Hz) (R, repetition rate) | 1000 | - | - |
| Pulse Length (s) (Tau or pulse duration) | 2.00E-08 | - | - |
| Linewidth (Hz) (Bandwidth) | 3.00E+09 | 300E+03 | 300E+03 |
| Linewidth (m) | 7.18E-12 | 7.18E-16 | 7.18E-16 |
| Spectral Irradiance (W/m3) | 6.96E+15 | 1.39E+15 | 2.79E+15 |
| Pct Spectral Irr. of original Pulsed Laser | | 20% | 40% |

Figure 11.    Pu 90% ionization relating to 40% CW laser power, *left* from [13]

## 1.    Availability, Cost and Feasibility

Market research of over six commercially available laser production companies identified that the $\lambda_1$ = 420.760 nm wavelength at the desired power of up to 2 Watts was not available as an off-the-self fiber optic laser product. Many of the fiber optic lasers can achieve the required power but their gain medium would not allow wavelengths in $\lambda_1$ or $\lambda_2$. The $\lambda_2$= 847.282 nm wavelength was available as a diode seed laser, but power outputs for a diode laser will not be sufficient for particle ionization.

Laser wavelength and power output requirements were the primary cost drivers. The traditional Ti-Sapphire laser with a wide tuning range discussed previously is also available as a CW laser, though the cost was near or greater than the current RIMS laser. The most cost-efficient solution was a COTS diode seed laser operating in the $\lambda_2$ wavelength with an external amplifier. Figure 12, shows the concept of the replacement of a single pulsed laser with a continuous laser (boxed in red).
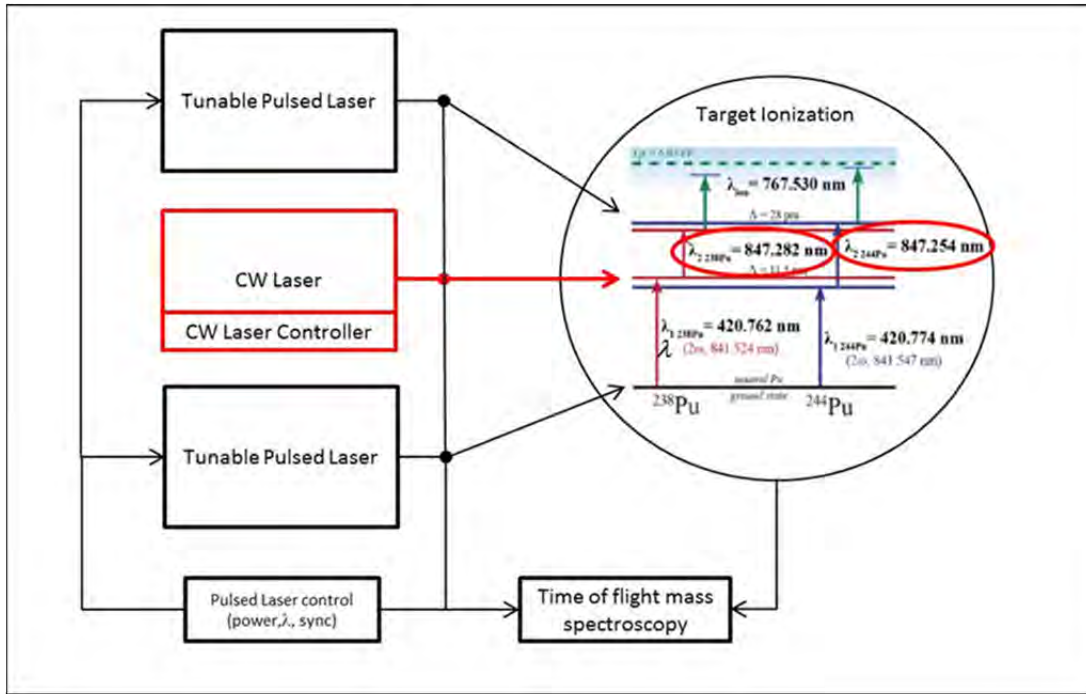
Figure 12.    Replacement of second pulsed laser with COTS CW laser

The feasibility considerations were how "plug-n-play" the CW laser would be while requiring little to no modifications. Many high power lasers are used in the welding and cutting industries, but modifications to focus and filter the energy would require the implementation of extensive and costly laser and optics specialization.

Taking all this into account and submitting an open bid to four qualified laser manufacturers, one company met the requirements to provide a 840–860 nm tunable wavelength laser, with power outputs >2 Watt through external tuning, and a tuning range of 20 nm. The laser was selected and purchased for evaluation.

## C.    DESCRIPTION OF THE SACHER LASER

The laser selected is the German built Sacher Lasertechnique TEC-420–0850–2000 CW Laser. The laser is a Master Oscillator Power Amplifier (MOPA) laser of the Littman/Metcalf design (see Figure 13). It is more complicated than a standard diode laser, but the additional complexity is necessary to provide the capability of boosting the power to the required 2 Watts through external amplification.
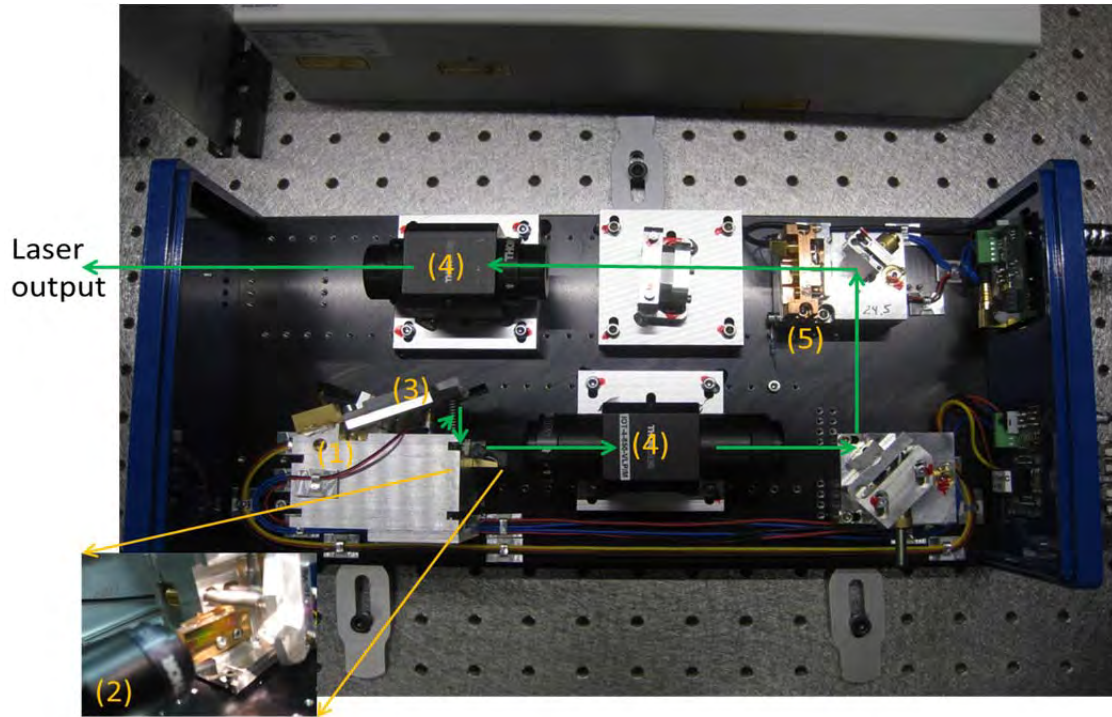
17

Figure 13.    Sacher Laser layout overhead view

The laser is designed to operate in the 840–860 nm range of wavelengths with a diode seed laser. Significant components that contribute to the gain, wavelength, and power output of the box are shown in Figure 13. They include the (1) seed laser (not actually shown), (2) diffraction grating, (3) reflection mirror, (4) optical isolators, and (5) laser amplifier. The entire COTS laser is enclosed in a space smaller than an average shoebox or about 3–4 times smaller than a single laboratory pulsed laser. Recall this is for only one of three lasers that the RIMS system will require; although smaller in footprint than the full system, it would still not be man-portable in this prototype configuration.

### 1.    Mode Control, Lasing and Wavelength Tuning

The Littman/Metcalf is a cavity design seed laser developed to maintain "mode-hop free" tuning through a pivot point technique in which the grating and the reflection mirror rotate about simultaneously [18], [19].

This design treats the grating and the reflective mirror as part of the lasing cavity (see Figure 14). When the seed laser diode is first powered up, a coherent but polychromatic light will strike the diffraction grating. The grating is angled such that the purely transmitted mode (m = 0) is directed outwards in a prescribed direction. The m = 1 (and greater) mode is light that is increasingly diffracted or spread. This light is directed towards a mirror [20]. From diffraction theory (Figure 15), the polychromatic light is spread out by wavelength and reflected back towards the grating (see Figure 16).
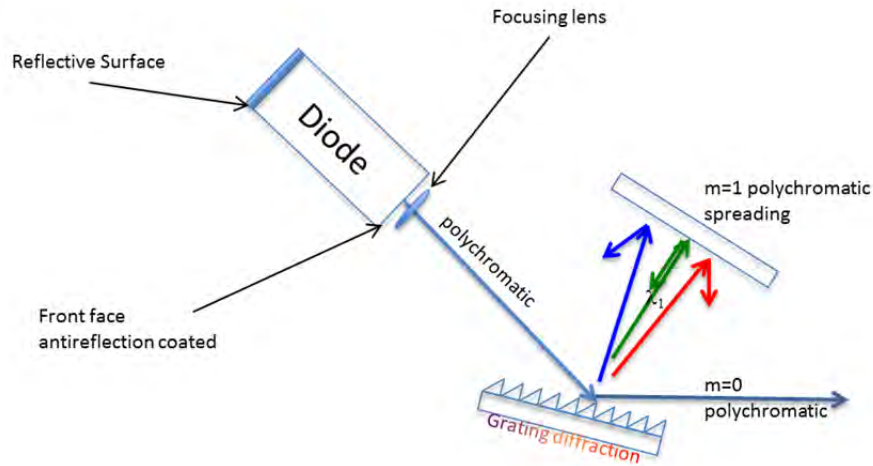


Figure 14.    Phase 1, initial laser power-up
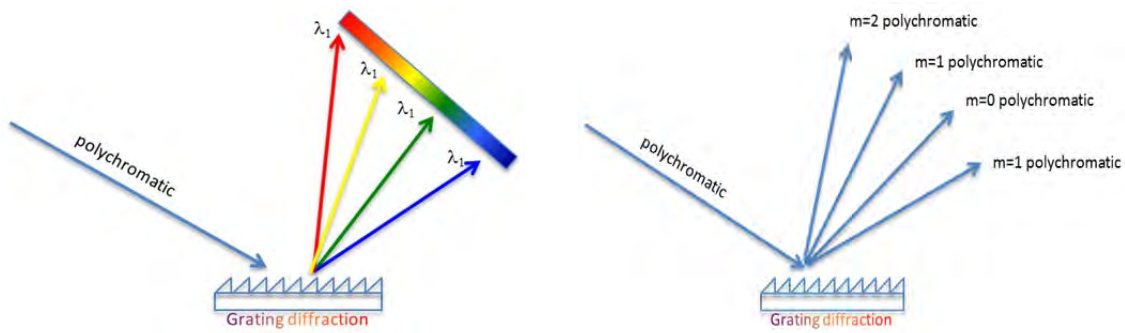


Figure 15.    *Left* - Standard diffraction , *Right* - Grating with "m" spreading
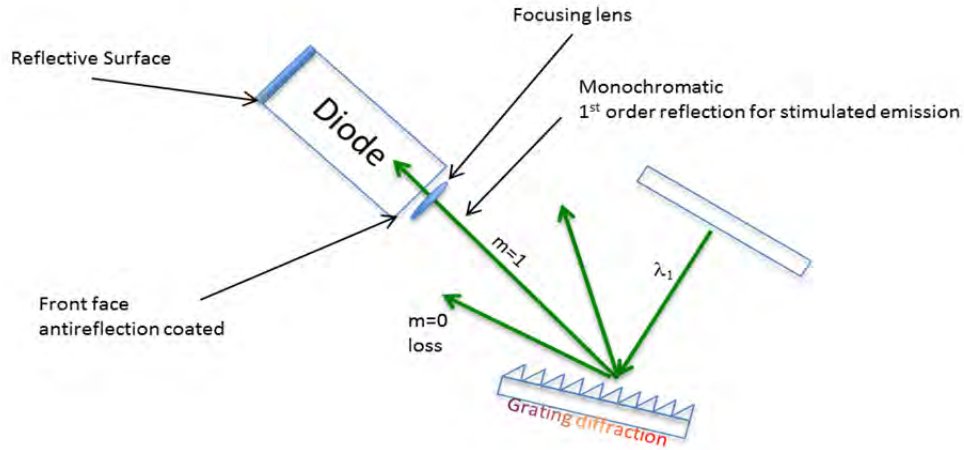
Figure 16.　Phase 2, monochromatic light only

The light reflected back to the diffraction grating is monochromatic and when it encounters the grating it further diffracts losing the m=0 mode and using the m=1 mode for stimulated emission (see Figure 17). The now stimulated emission is monochromatic and again is m=0 diffracted by the grating. The resulting emission is used as the seed laser output.
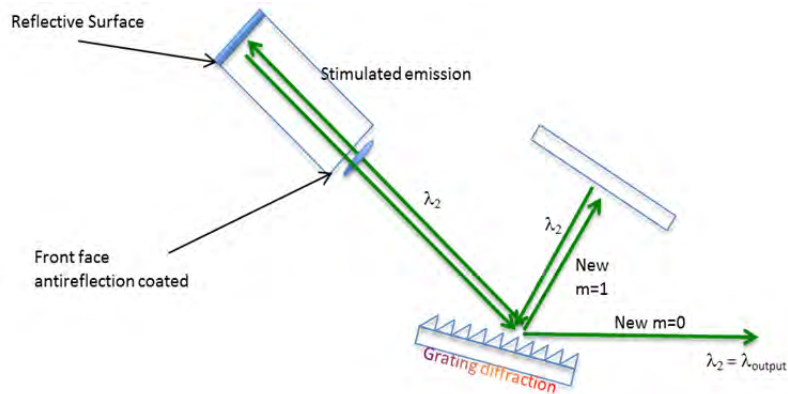


Figure 17.　Phase 3, sustained stimulated monochromatic emission

The design described is the method the Sacher Laser uses to achieve a single mode and wavelength tuning for the laser. A limitation of this design is its low lasing power output due to m=0 loss [21] from phase 2 as earlier described. For most

applications, including the RIMS application, to achieve higher output an amplifier is used [21], [22], [23].

## 2.        The Tapered Amplifier

Generally a diode seed laser provides outputs in milliwatts, a power not sufficient for the requirements to induce sufficient ionization. The method the Sacher Laser uses to boost this power is a tapered laser amplifier (see Figure 13, item 5). A tapered amplifier (see Figure 18) is fabricated similar to a semiconductor chip through different optically indexed materials. This is an optical amplifier that does not need to be converted into an electrical signal for amplification. When a tapered amplifier is used in a MOPA configuration it preserves the spectral properties of the seed laser (wavelength, linewidth and polarization) [21].
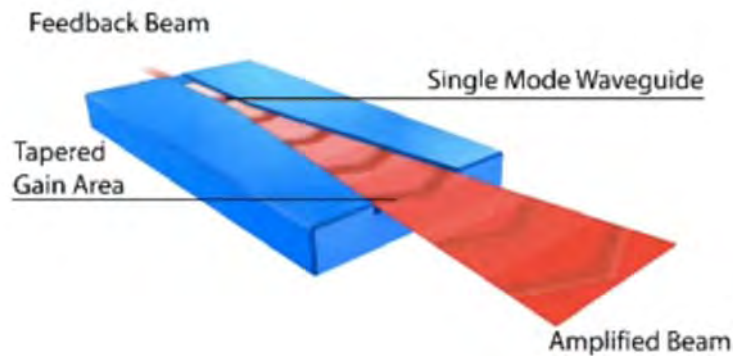


Figure 18.    Tapered amplifier design, from [21]

There are different types of tapered amplifiers they all have a narrow input followed by a widening output. The refractive index controlled material ($n_r$) is greater than the substrate material with a refractive index ($n_s$). The laser enters the input section with material $n_r$ and is internally reflected through the waveguide. This type of amplifying material is AlGaAs and it creates electron hole pairs (similar to the seed diode laser) for stimulated emission. The angle and design of the amplifier achieves the desired power gain while maintaining the seed lasers characteristics. Additional details of the operation of specific amplifier designs can be found in the article in Reference [24].

An important limitation of tapered amplifier is their susceptibility to overheating. They can overheat from high currents within the amplifier when it is powered without a seed laser [18]. The seed laser provides an optical outlet for the amplifier whereas otherwise the input current is converted to heat. Additionally, any backward propagation will focus to a high intensity at the narrow input heating up the chip [24]. To prevent this optical isolators are necessary.

### 3.  Optical Isolator

An optical isolator is a magneto-optic device that only allows light to travel in one direction preventing damage to the instruments, or interference to desired laser properties such as modes or wavelengths. The *Faraday Effect* rotates (β) a beam of linearly polarized light preserving its linearity on exit (see Figure 19).
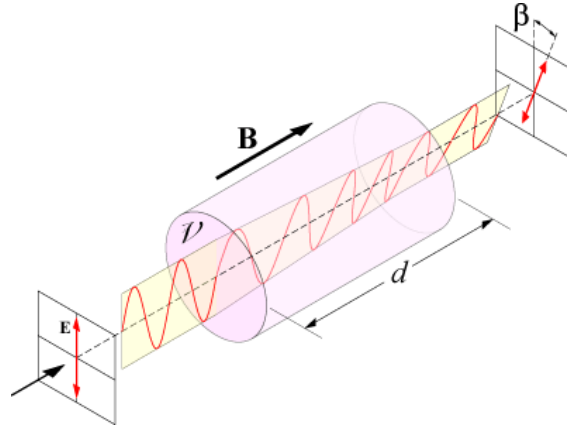


Figure 19.    Faraday Effect, from [25]

The dependence of the rotation is based on the equation

$$\beta = VBd ,$$

where *V* is the *Verdet constant* for the material, *B* is the magnetic field strength, and *d* is the length of the material. In the "forward mode" the laser enters a polarizer and becomes polarized, as it progresses through the *Faraday* rotator it rotates to a desired angle (β) in the clockwise direction, then it exits through another polarizer at angle β. In "reverse mode" or backscattering light, the beam enters the exit polarizer becoming polarized at β,

it enters the *Faraday* rotator and is rotated an additional angle β totaling a 90° rotation from the incoming light; as it encounters the entry polarizer it is now perpendicular to it thus being absorbed [8], [26].

Now with the above understanding of the inner workings of the COTS CW laser and the insight that it is capable of producing a continuous wave, single mode monochromatic laser beam, the characteristics of the laser in terms of power, thermal stability, "Gaussian" shape, wavelength, and linewidth was investigated through preliminary experiments as described in the next chapter.

THIS PAGE INTENTIONALLY LEFT BLANK

## IV. EXPERIMENT

### A. POWER, BANDWIDTH, LINEWIDTH AND BEAMSHAPE

As previously discussed sufficient laser power is necessary to produce required irradiance onto the target atoms. "The aim is to maximize the probability of ionizating the target isotope and minimize the ionization of other species." [2]. Required power of 2 Watts to ensure 90% target element ionization is necessary and introduced previously in, Figure 11. To ensure sufficient power is available for ionization, the CW laser power output and stability needed to be tested.

Sufficient irradiance is not enough to ensure ionization; the wavelength must also be tuned to that specific desired isotope. Experimental results from previous RIMS research (Figure 20) on Uranium show that a difference of 5 pm can result in an ionization preference for either $^{235}$U or $^{238}$U. "Broad" and "narrow" bandwidth testing shows the selectivity and sensitivity of wavelength and linewidth.
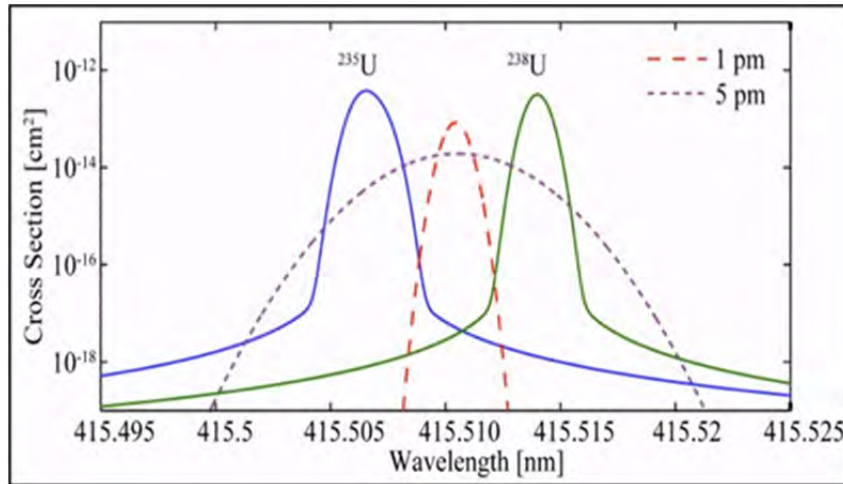


Figure 20.    RIMS result with laser linewidths of 1 and 5 pm, from [13]

Experimental results additionally show that narrower bandwidth lasers leads to greater sensitivity and selectivity between isotopes (Figure 21).
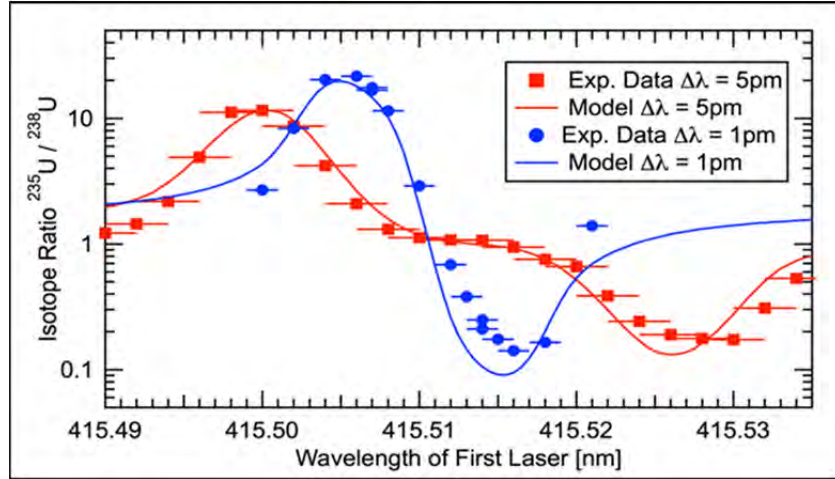
Figure 21.    Experimental RIMS results showing greater sensitivity with narrower linewidth, from [13]

The beam shape characteristic is important to ionization because spatial distribution must ensure sufficient intensity throughout the ionization target. A Gaussian beam profile and a beam quality factor ($M^2$) gives information on the intensity distribution, beam divergence, and beam size. Such information can be used for placement and focusing of the beam onto the ionization target. The previously discussed laser characteristics are necessary in understanding and achieving ionization.

## 1.    Test Plan

Testing and evaluation of the Sacher laser was conducted to determine and verify the output power, power/temperature stability, beamshape, wavelength and linewidth (see Table 1). It was conducted at Lawrence Livermore National Laboratory using the same equipment used in the existing RIMS project for laser operational testing. Only initial evaluations were completed due to time constraints. Although these initial evaluations are not exhaustive, they give insight into the requirements for calibration and identification of future testing needs.

Table 1.     COTS laser test plan

| Test Plan | |
|---|---|
| Short Time (Pulse to Pulse) & Long Time (Hrs/Days) | |
| | Method/Instrument |
| | |
| Measure Mean Wavelength | WS-6-200 (IR 800-1750 nm) |
| Measure Power | Gentec-EO (1uJ or uW) |
| Measure Beamshape | Data Ray Beam Profiler (Gaussian shaped?) |
| Wavelength (Tunability) | Self Heterodyne |
| | Dual Heterodyne |
| | Interferometer |

## B.     EXPECTED AND VERIFIED CHARACTERISTICS

Preliminary safety checks of the laser provided satisfactory results, and the laser was slowly powered up to prevent damage to the tapered amplifier. All readings were conducted on the same day with environmental temperature stable. The initial testing and evaluation were conducted at full power up to 45 minutes. Future stability studies should include additional long term and overnight tests.

### 1.     Seed Current and Output Power

The output power of the laser is of great importance because as discussed in Chapter III and Figure 11 the desired power to achieve 90% particle ionization is ~ 2 Watts of a CW laser. Figure 22, is a picture showing the power output of the laser measured though a Gentec-E Maestro power meter at 1.94 Watts.
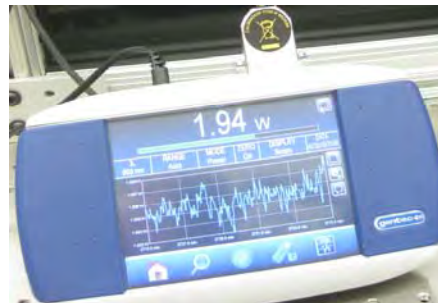


Figure 22.    Sacher laser power output reading (time variation output provided)

Figure 23 shows the current input to the seed laser at 100 mA and into the tapered amplifier at 4000 mA both at their maximum at this output power.



Figure 23.    Seed and Amplifier max current settings for 1.94 Watt laser output

Figure 24 is a plot of the Output Power vs. Seed Current as measured at ~847nm up to 1.94 W. This plot and the expected output power is similar in shape to the manufacturer provided plot with a slight degradation in power. Degradation can be attributed to slight misalignments from the transport of the laser and is being verified with the manufacturer.



Figure 24.    COTS Output power vs. Input current compared to manufacturer specification

## 2. Power and Temperature Stability

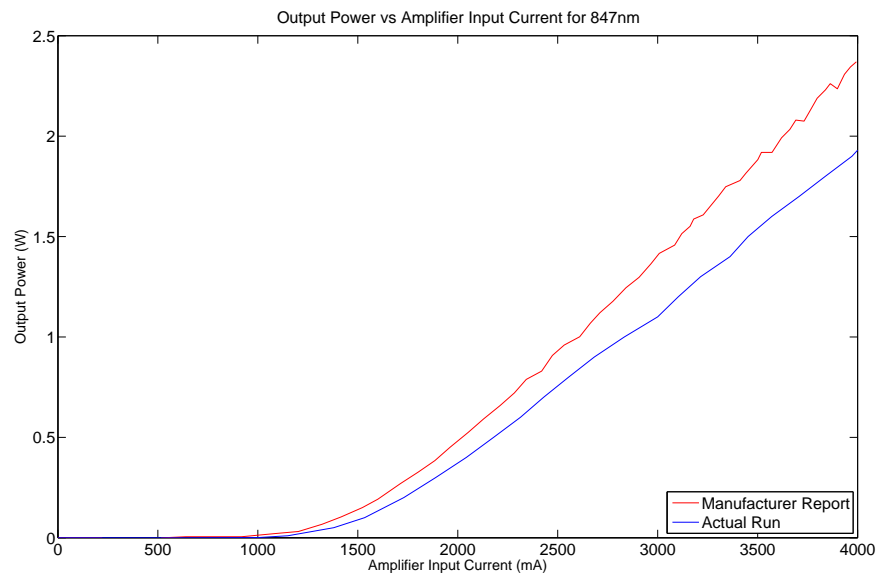The power and temperature stability of the Sacher laser is important to verify and understand. As discussed previously varying temperatures have effects on the wavelength due to cavity and grating misalignments. Encouragingly, once both the seed laser and amplifier achieved operating temperature at ~30 mins they held steady through several hours of testing at 22°C and 19°C, respectively.

The power stability measured at 30 minutes showed a fluctuation from mean of ~ 0.02 mW Figure 25 shows a Power vs. Time plots with the fluctuations similar in scale. Figure 26 shows a relatively small standard deviation of ~0.004 W. The different power levels can be attributed to slight misalignments of the Gentec power meter between different experimental setups.
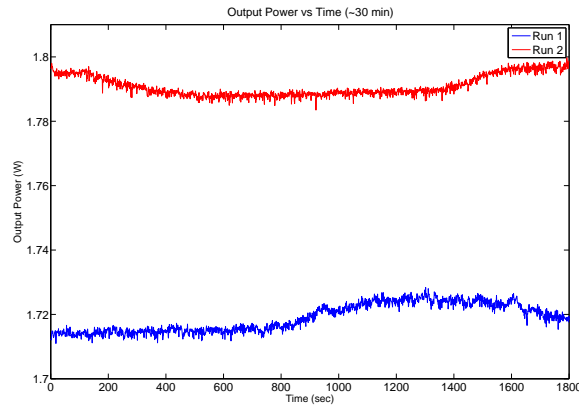


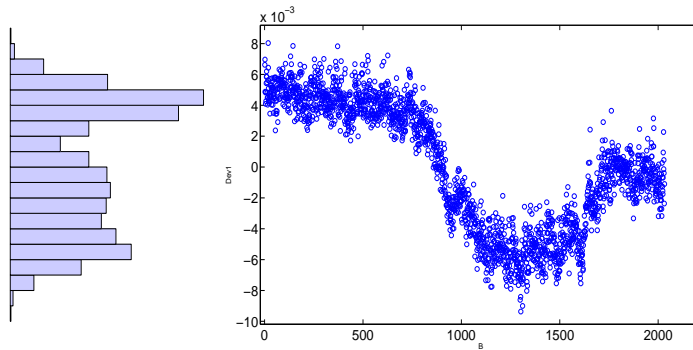Figure 25.    Different runs with power meter positioned differently



Figure 26.    Run 1, histogram showing peak standard deviation ~0.004 W

29

### 3.     Beamshape

The beamshape is of particular interest because as discussed in Chapter II Section 3, a Gaussian shaped or $TEM_{00}$ beam is preferred. Multimode lasing can cause deviations from a Gaussian. It was necessary to beam split the laser output to ~10% power to prevent damage to the profiler.

A Data Ray Profiler was used to take the readings in Figure 27; it can be subjectively observed that the beam appears single mode when compared to the $TEM_{00}$ profiles in Figure 9. There is a slight second peak observable to be approximately 1/10 the intensity of the main beam; it can be attributed to another mode, but further testing is needed for determination.
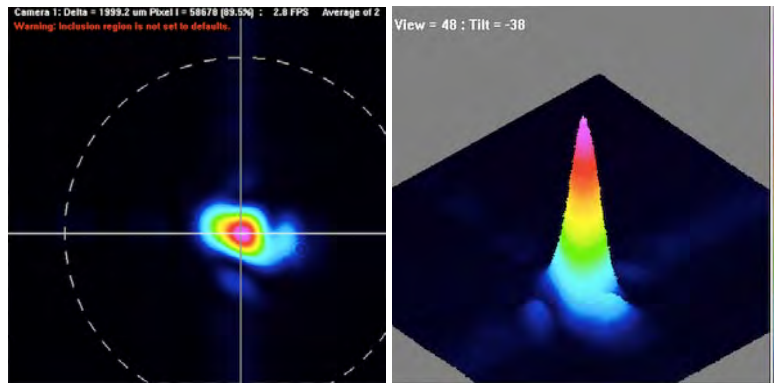


Figure 27.     Data Ray Profile screen capture pictures

Figures 27 shows a fairly Gaussian profile (red) when overlaid to the power output in black. The profile is showing a Gaussian fit of 90% in the axis with the additional peak and 97% on the orthogonal axis
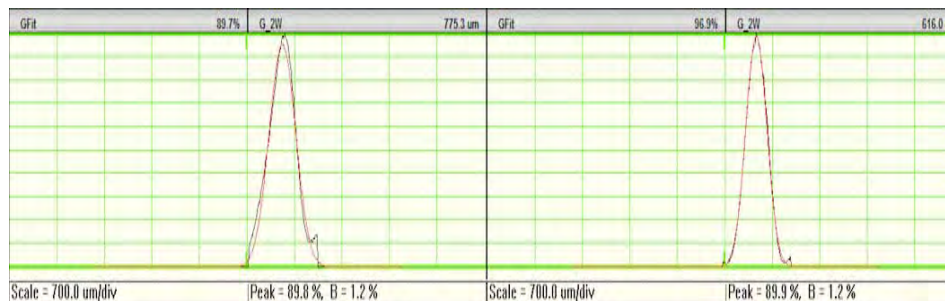


Figure 28.     Orthogonal axis comparison for laser profile to Gaussian curve

Additional measurements need to be taken to account for the beam divergence and focal lengths to confirm the beam quality factor ($M^2$) used in industry. It is defined by ISO/DIS 11146 and determines the Gaussian fit related by the equation

$$\theta_0 = M^2 \frac{4\lambda f}{\pi D_0},$$

where $\theta_0$ is the beam divergence angle and $D_0$ is the beam waist width. A $M^2$ value close to 1 is preferred; however, $M^2$ is proportional to power output and should be considered as a tradeoff. To conduct a $M^2$ fit several measurements will need to be made at different focal lengths [27].

### 4. Wavelength and Linewidth

The wavelength and the linewidth measurements proved to be the most challenging. The instruments available on-hand were a WS-6 and Ocean Optics HR2000. Both of these instrument measure linewidths to about a picometer scale and are not intended for measurements to femtometers scale (the expected linewidth). "Typical grating based optical spectrum analyzers do not offer the measurement resolution required for very narrow linewidths" [28].

Through preliminary measurements, the wavelength was found to be ~847+/- 1 nm (Figure 28). This variability would be too large to use for Pu ionizations. The manufacturers specification shows a very narrow linewidth of <100kHz (~0.24 fm) and further calibration and testing of the wavelength and linewidth is needed to confirm this resolution.
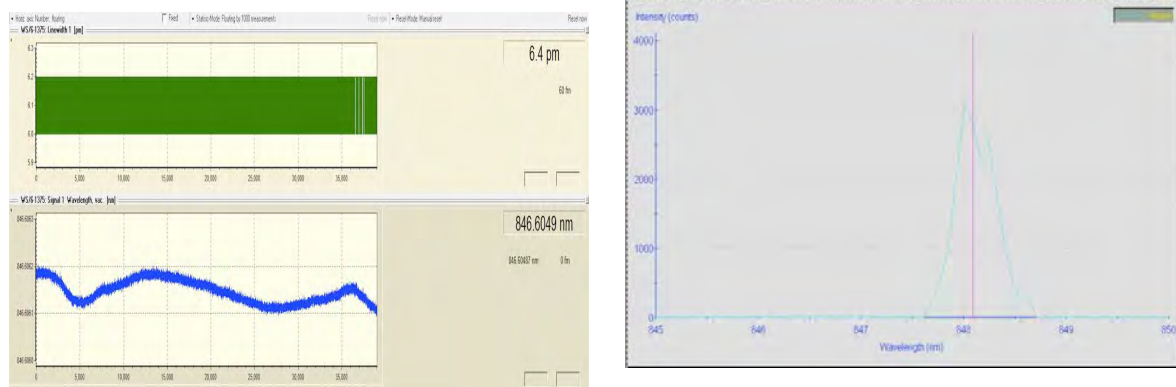
Figure 29.    *Left-* WS6 Wavelength and linewidth measurements.
*Right-* Ocean Optics wavelength and linewidth measurements

The manufacturer used a heterodyne method to measure the linewidth. Two such methods are: dual heterodyne which uses either a known calibrated reference laser of narrower linewidth, or delayed self-heterodyne which uses an interferometer to delay one of the split paths. Both methods measure the frequency difference between the reference and test beam at a photodiode. The heterodyne technique makes it possible to calculate the linewidth of external cavity lasers on the order of kilohertz linewidths [29].

# V.   CONCLUSION/RECOMMENDATIONS

In this thesis research, the potential to replace existing excitation lasers for the RIMS nuclear forensics application was investigated. The effort began with a feasibility study concluding that it would be possible to implement and where technical, operational and cost advantages could result from doing so.

This led to the follow-on effort to identify and procure a candidate COTS CW laser system for further evaluation of the concept. From characterization of the attributes of available COTS lasers, it was possible to identify and procure an acceptable candidate CW laser that could be used to replace a pulsed laser in RIMS.

The Sacher Lasertechnique TEC-420–0850–2000 CW Laser was acquired and testing was conducted to initiate the characterization of the laser and further explore its potential suitability for the RIMS nuclear forensics application.

The continuous wave laser manufactured by Sacher Lasertechnik and selected for the RIMS project shows great potential as a replacement candidate for the Ti-Sapphire laser in the second ionization of plutonium. At a fraction of the cost and significant size reduction, the continuous laser could potentially replace two or possibly all three of the pulsed lasers used for stepwise excitation to ionization of target elements. The COTS laser was found to have an expected wavelength range of approximately 20 nm in preliminary tests, but that needs verification. Such a laser has the potential to be suitable for both uranium and plutonium ionization as discussed in Chapter I.

Future work should focus on experiments making reliable measures for the wavelength and the linewidth, using methods such as the hetrodyne techniques discussed previously. With knowledge of the linewidth wavelength tuning can be further investigated. The COTS laser has a manual adjustable wavelength tuning screw that can be automated with an additional attachment if necessary.

Furthermore, actual ionization experiments should be conducted to verify satisfactory power requirements and stability. The beamshape can also be studied to test for ionization and whether the slight deviation from Gaussian impacts ionization.

THIS PAGE INTENTIONALLY LEFT BLANK

# LIST OF REFERENCES

[1]     Nuclear Forensics. (2015, Jan. 1). Wikipedia. [Online] Available: http://
        http://en.wikipedia.org/wiki/Nuclear_forensics.

[2]     B. H. Isselhardt, "Quantifying uranium isotope ratios using resonance ionization
        mass spectrometry: The influence of laser parameters on relative-ionization
        probability," Lawrence Livermore Laboratory, Livermore, CA, LLNL-TH-
        500737, Sept 2011.

[3]     G. Hurst, M. Payne, S. Kramer, and J. Young, "Resonance ionization
        spectroscopy and one atom detection," *Rev. Mod. Phys.,* vol.51, no. 4 pp. 767–
        819, Oct. 1979.

[4]     J. Young, R. Shaw, and D. Smith, "Resonance ionization mass spectrometry,"
        *Anal. Chem.*, vol. 61, no. 22 pp. 1271–1279, Oct. 1979.

[5]     K. Knight, M. Savina, S. Prussin, and I. Hutcheon, "Multi-actinide isotopic
        measurements from a single sample by resonance ionization mass spectrometry,"
        presented at LLNL- Conf 486708, 2011.

[6]     How do LEDs Work? (2015, May 15). Embedded adventures. [Online].
        Available: http://www.embeddedadventures.com/Tutorials/tutorials_detail/118.

[7]     S. O. Kasap, *Optoelectronics and Photonics Principles and Practices*, 2nd ed.
        New York City, NY: Pearson Education Ltd, 2013, pp. 242–247.

[8]     F. L. Pedrotti and L. S. Pedrotti, *Introduction to Optics,* 2nd ed. Upper Saddle
        River, NJ: Prentice Hall, 1993, pp. 251–254, 260, 440–446, 555.

[9]     R. S. Quimby, *Photonics and Lasers An Introduction*, Hoboken, NJ: John Wiley
        & Sons Inc, 2006, pp. 159–162, 198–199, 207–208, 314–317, 381–389, 397–402,
        440.

[10]    Stimulated Emission Devices LASERS. (2015 Mar. 13). National Instruments.
        [Online] Available: http://www.ni.com/white-paper/14878/en/.

[11]    Laser. (2015, Feb. 1). Wikipedia. [Online] Available:
        http://en.wikipedia.org/wiki/Laser.

[12]    Lasers. (2015, Apr. 4). Coherent Inc. [Online] Available:
        http://www.photonics.com/EDU/Handbook.aspx?AID=25161.

[13] B.H. Isselhardt, M. R. Savina, K. B. Knight, M. J. Pellin, I. D. Hutcheon, and S. G. Prussin, "Improving precision in resonance ionization mass spectrometry: influence of laser bandwidth in uranium isotope ratio measurements," *Anal. Chem.*, vol. 10, no. 1021 pp. 2469–2475, Mar. 2011.

[14] Multi-mode Optical Fiber. (2015, Mar. 1). Wikipedia. [Online] Available: http://en.wikipedia.org/wiki/multimode_optical_fiber.

[15] J. Hecht, *The Laser Guidebook*, 2nd ed, Blue Ride Summit, PA: Mc Graw Hill Professional, 1999, pp. 101–109.

[16] F. Dominec. (2015, Mar 12). File: Lasergaussianprofile.svg, Wikimedia. [Online] Available: http://commons.wikimedia.org/wiki/File:Laser_gaussian_profile.svg.

[17] Longitudinal and Transverse Modes. (2015, Mar. 15). OPI. [Online] Available: http://www.optique-ingenieur.org/en/courses/OPI_ang_M01_CO3/Contenu_02.html.

[18] P. McNicholl, and H. Metcalf, "Synchronous cavity mode and feedback wavelength scanning in dye laser oscillators with gratings," *App. Optics*, vol. 24, no. 17 pp. 2757–2761, Sept. 1985.

[19] K. Liu, and M. Littman, "Novel geometry for single-mode scanning of tunable lasers," *Opt. Soc. of Amer.*, vol. 6, no. 3 pp. 117-118, Nov. 1980.

[20] Diffraction Grating. (2015, Feb. 5). Wikipedia. [Online] Available: http://en.wikipedia.org/wiki/Diffraction_grating.

[21] Principles of Mode-Hop Free Wavelength Tuning No.13. (2015, May 1). Sacher Lasertechnik. [Online] Available: https://secure.sacher-laser.com/downloads/-technical_documents/t03d6w-en.pdf.

[22] J. H. Marquardt, F. C. Cruz, M. Stephens, C. W. Oates, L. Hollberg, and J. C. Bergquist, "Grating-tuned semiconductor MOPA lasers for precision spectroscopy," *SPIE*, vol 2834, no. 34 pp. TN71–77, Oct. 1996.

[23] J. Sacher, private communication, May 1, 2015.

[24] Y. Xiong, S. Murphy, and J. L. Carlsten, "Design and characteristics of tapered amplifier diode system by seeding with continuous-wave and mode-locked external cavity diode laser," *Opt. Engr.*, vol 45, no. 12 pp. 124205–1, 214205–5, May 2006.

[25] B. Mellish. (2015, Apr. 30). File: Faraday-effect.svg, Wikimedia. [Online] Available: http://commmons.wikimedia.org/wiki/File:Faraday-effect.svg

[26]    Optical Isolator Tutorial. (2015, May 1). Thorlabs. [Online] Available:
        http://www.thorlabs.us/rewgrouppage9.cfm?objectgroup_id-4914.

[27]    Laser beam quality (2015, Apr.10, 2015). Wikipedia. [Online] Available:
        http://en.wikipedia.org/wiki/Laser_beam_quality.

[28]    X. Chen, "Ultra-narrow laser linewidth measurement," Ph. D. dissertation, Dept.
        Elect. Engr., Virginia Polytech., Blacksburg, VA, 2006.

[29]    H. Ludvigsen, M. Tossavainen, and M. Kaivola, "Laser linewidth measuremetns
        using self-homodyne detection with short delay," *Optics Comms.*, vol. 155, no. 1
        pp. 180–186, Oct 1998.

THIS PAGE INTENTIONALLY LEFT BLANK

# INITIAL DISTRIBUTION LIST

1.      Defense Technical Information Center
        Ft. Belvoir, Virginia

2.      Dudley Knox Library
        Naval Postgraduate School
        Monterey, California